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FULL LENGTH ARTICLE

Simple optimization method for partitioning purification of hydrogen networks

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Abstract The Egyptian petroleum fuel market is increasing rapidly nowadays. These fuels must be in the standard specifications of the Egyptian General Petroleum Corporation (EGPC), which required lower sulfur gasoline and diesel fuels. So the fuels must be deep hydrotreated which resulted in increasing hydrogen (H_2) consumption for deeper hydrotreating. Along with increased H_2 consumption for deeper hydrotreating, additional H_2 is needed for processing heavier and higher sulfur crude slates especially in hydrocracking process, in addition to hydrotreating unit, isomerization units and lubricant plants. Purification technology is used to increase the amount of recycled hydrogen. If the amount of recycled hydrogen is increased, the amount of hydrogen that is sent to the furnaces with the off gas will decrease. In this work, El Halwagi et al. (2003) and El Halwagi (2012) optimization methods which are used for recycle/reuse integration systems have been extended to be used in the partitioning purification of hydrogen networks to minimize the hydrogen consumption and the hydrogen discharge. An actual case study and two case studies from the literature are solved to illustrate the proposed method.

Abbreviations: DHT, diesel hydrotreating unit; FHP, fresh hydrogen plant flowrate; HCU, hydrocracking unit; ISOM, isomerization unit; NHT, naphtha hydrotreating unit; PSA, pressure swing adsorption; $\text{Cost}_{\text{operating}}$, operating cost; OC_{H_2}, hydrogen production operating cost; OC_{Fuel}, fuel gas value operating cost; OCH, cost of production; OCF, heat cost of fuel; \$/MBTU, dollar for each million BTU; i, j, from source i to sink j; F_{Fuel}, fuel flow rate; F_{in}, inlet flow rate to purifier; F_j, flow rate of fresh hydrogen to sink j; F_{product}, flow rate of product stream of the purifier; F_{residual}, flow rate of residual stream of the purifier; LHV_{H_2}, lower heating value of hydrogen; LHV_{CH4}, lower heating value of methane; N_{sinks}, total number of sink streams; N_{sources}, total number of source streams; R, hydrogen recovery of the purifier; w_i, flow rate of source stream; $w_{i,j}$, flow rate from source i to sink j; y_{in}, impurity concentration in the feed stream to the purifier; y_{product}, composition of product stream of the purifier; y_{residual}, composition of residual stream of the purifier; z_j^{in}, composition of sink j after mixing of sources fractions and fresh fractions; z_j^{max}, maximum allowable impurity concentration of sink j; z_j^{min}, minimum allowable impurity concentration of sink j

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1. Introduction

The research regarding refinery hydrogen management can trace its history back to 1980s. But hydrogen usage has increased day by day, due to many new factors. First, stricter legislation on sulfur and aromatic levels in petroleum fuels increases the need for hydrotreating to produce low sulfur fuel products [1]. Second, the shift toward processing heavier crude oils and the reduction in the demand for heavy fuel oil is forcing greater use of hydrocracking processes for upgrading heavy oils to middle distillates [2]. One of the methods to minimize the hydrogen usage in refinery is that, Hydrogen is rich in off-gas of many processes such as delayed coking and catalytic cracking. However the purity of off-gas may not be high enough. So sometimes a purifier is introduced to improve the concentration of hydrogen before sending it to a hydrogen consumer. Purifiers can also remove hazards and impurities in hydrogen, which allows hydrogen consumers to use it in a more efficient way [3].

The main hydrogen purification technologies used in refineries are pressure-swing adsorption (PSA), selective permeation using polymer membranes, and cryogenic separation. These purification units can be used to remove impurities and increase the hydrogen concentration. These recovery technologies are based on different separation theories. Hydrogen concentration, impurity characteristics and available pressure of off-gas determine the selection criteria [3].

Over the past decade, several design techniques have been developed to minimize hydrogen usage in refinery plant through efficient process integration and maximizing recycle/reuse [4].

The problem of synthesizing mass exchange networks (MENs) has been first introduced by El-Halwagi and Manousiouthakis [5]. It seeks to transfer certain species from a set of rich streams to a set of lean streams. This problem is referred to as the recycle/reuse problem. The objective function of the recycle/reuse problem is to allocate various process sources (or streams) to process sinks (units that can employ the sources) so as to minimize the consumption of the fresh resource as hydrogen [4]. Alves and Towler [6] have proposed a systematic method for the analysis of hydrogen distribution systems based on the concept of hydrogen surplus. Their method sets targets for the minimum flow rate of fresh hydrogen required by the refinery before any system design. The analysis method is used to provide quantitative insights and to identify the existence of bottlenecks in the hydrogen distribution system. El Halwagi et al. [4] developed a rigorous and non-iterative graphical method to minimize the fresh resource consumption. Foo and Manan [7] put forward a numerical targeting method, named the gas cascade analysis (GCA) to calculate the utility target. Zhao et al. [8] take into account impurities concentration within a hydrogen network. Liu and Zhang [3] developed an automated design superstructure approach that demonstrates the choice of purifier selection as well as their integration in the hydrogen networks. The objective function for the mixed integer non linear program problem could be minimum hydrogen utility, operating costs or the total annualized cost of the network. Fonseca et al. [9] proposed a linear programming (LP) method to solve refinery hydrogen network optimization problems. The authors utilized the simplified hydrogen consumer model developed by Alves [10] and constructed an LP formulation in terms of mass balance between sinks and sources under pressure consideration. Jia and Zhang [11] introduced a more realistic approach to multi-component optimization of refinery hydrogen network by assuming constant vapor–liquid equilibrium ratios for slight changes in the flash inlet stream composition.

Tahouni et al. [12] proposed a new optimization mathematical model for hydrogen management in petrochemical complexes based on setting a comprehensive superstructure model. This superstructure including purifier and compressor of hydrogen plant or catalytic reformer unit.

Shariati et al. [13] presented a comprehensive analysis which is carried out on petrochemical units using a modified automated targeting technique. The modified automated targeting technique is applied to determine the minimum hydrogen consumption.

In this work, a simple optimization method for partitioning purification systems of hydrogen networks is proposed as an extension of El Halwagi et al. method [4] and El Halwagi method [14] which are used for recycle/reuse systems. Partitioning purification systems are used to minimize the hydrogen consumption and the hydrogen discharge in refinery. This method determines which source from the existing sources would be used for the purification. One partitioning purification unit would be used.

The formulation is a linear program that can be solved globally by LINGO program V.11.

2. Problem statement

For a given process there is a set of process sinks and a set of process sources described as follows:

- The set of process sinks: SINKS = $\{j|j = 1, 2, \ldots, N_{\text{sinks}}\}$. Each sink requires a feed with a given flowrate, G_j, and a composition of a single targeted species, z_j^{in}.
- The set of process sources: SOURCES = $\{i|i = 1, 2, \ldots, N_{\text{sources}}\}$, can be recycled/reused in process sinks. Each source has a given flow rate, w_i, and a given composition, y_i.
- Also available for service is a fresh (external) resource that can be purchased to supplement the use of process sources.
- There is a purification unit that is used to decrease the targeted species from the sources.

First the problem is analyzed to allocate sources to sinks only, and then is expanded to the options of process purification of streams to satisfy the demands of the sinks at minimal fresh resource.

3. Problem representation

(1) The first step in the analysis is to represent the problem through a source–sink representation as described by El Halwagi et al. method [4] as shown in Fig. 1 to get the minimum fresh hydrogen and minimum hydrogen discharge without any purification unit.

The objective function is to minimize the fresh hydrogen

$$\text{Minimum consumption of fresh hydrogen} = \sum_{j=1}^{N_{\text{sinks}}} F_j \tag{1}$$

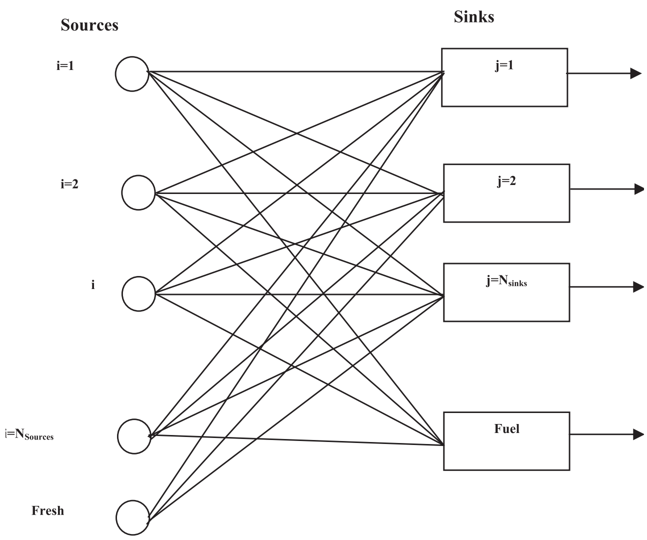

Figure 1 Source/sink allocation.

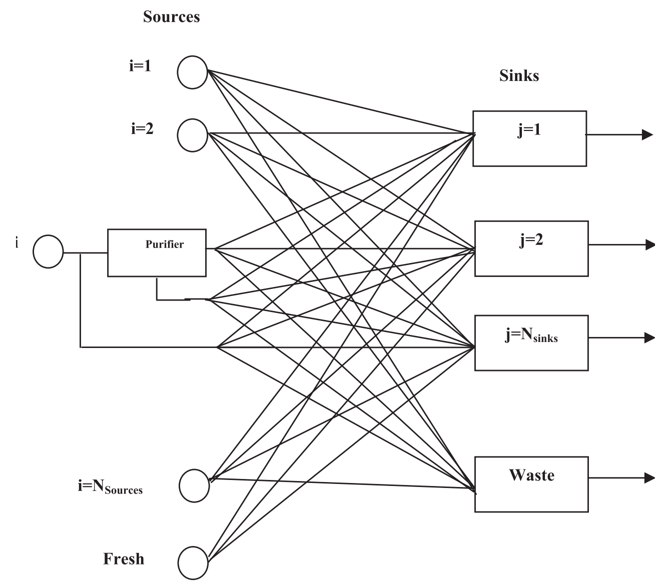

Figure 2 Source/sink allocation with purification unit.

Each source i, is split into N_{sinks} fractions as described in Fig. 1. The flowrate of each split is expressed as $w_{i,j}$. Also, one split is forwarded to the fuel sink which is denoted by $w_{i,\text{Fuel}}$:

$$w_i = \sum_{j=1}^{N_{\text{sinks}}} w_{i,j} + w_{i,\text{Fuel}} \quad \text{for } i = 1, 2, \ldots, N_{\text{sources}} \tag{2}$$

The following step is the mixing of the split fractions into a feed to the jth sink. The split fractions come from the process sources and the fresh stream:

$$G_j = F_j + \sum_{i=1}^{N_{\text{sources}}} w_{i,j} \quad \text{for } j = 1, 2, 3, \ldots, N_{\text{sinks}} \tag{3}$$

where F_j is the amount of fresh hydrogen sent to the jth sink:

$$G_j z_j^{\text{in}} = F_j x_f + \sum_{i=1}^{N_{\text{sources}}} w_{i,j} y_i \quad \text{for } j = 1, 2, 3, \ldots, N_{\text{sinks}} \tag{4}$$

where x_f is the impurity concentration of the fresh hydrogen.

$$z_j^{min} \le z_j^{in} \le z_j^{max} \quad \text{for } j = 1, 2, \ldots, N_{\text{sinks}} \tag{5}$$

$$w_{ij} \ge 0 \quad \forall i = 1, 2, \ldots, N_{\text{sources}}, \quad \forall j = 1, 2, \ldots, N_{\text{sinks}} \tag{6}$$

$$F_j \ge 0 \quad \forall j = 1, 2, \ldots, N_{\text{sinks}} \tag{7}$$

(2) The second step is to represent the problem as a process purification of streams by adding a purification unit to source i as described in Fig. 2. Pressure swing adsorption (PSA), the membrane, and the cryogenic separation are the common types of hydrogen purifiers [7,15]. The hydrogen purification units separate single hydrogen feed stream into top product stream and residual product stream. The top product stream has a lower impurity concentration than the residual stream. The top product stream can be reused or recycled in the hydrogen network but the residual stream can be purged or used as fuel [7,15].

To determine the location of the purifier, it is assumed that all sources are purified. For each source i, an amount equal to the hydrogen discharge is purified and the rest of source i is integrated in the network as described in Fig. 2. We have different cases for the location of the purifier equal to the number of the sources. Each case has two new sources (product and residue of the purifier) and modified source i. All sources flowrates are the same except source i at which the purifier is placed. The flowrate of modified source i is an old value of it before placing the purifier minus the discharge flowrate determined by the first step. For each case we solve the network to obtain the new fresh hydrogen and the new hydrogen discharge using Eqs. (8)–(10) for the mass balance around the purifier [15].

$$F_{\text{in}} = F_{\text{product}} + F_{\text{residual}} \tag{8}$$

$$F_{\text{in}} y_{\text{in}} = F_{\text{product}} y_{\text{product}} + F_{\text{residual}} y_{\text{residual}} \tag{9}$$

$$R F_{\text{in}} y_{\text{inH}_2} = F_{\text{product}} y_{\text{productH}_2} \tag{10}$$

where R is the hydrogen recovery.

(3) The third step is to calculate the annual operating cost for the hydrogen network with each purifier as follows:

$$\text{Cost}_{\text{operating}} = \text{OC}_{\text{H}_2} - \text{OC}_{\text{Fuel}} \tag{11}$$

$$\text{OC}_{\text{H}_2} = \text{OCH} * \text{FHP} \tag{12}$$

OC_{H_2} represents the hydrogen production operating cost which is a function of the FHP hydrogen plant flowrate, multiplied by the cost of production, OCH.

$$\text{OC}_{\text{Fuel}} = \text{OCF} * F_{\text{Fuel}} * (\text{LHV}_{\text{H}_2} * y_{\text{H}_2} + \text{LHV}_{\text{CH}_4} * y_{\text{CH}_4}) \tag{13}$$

OC_{Fuel} represents the fuel gas value operating cost. Assuming that the fuel gas is a mixture of hydrogen and methane, fuel gas value operating cost is function of the summation of fuel gas heating value LHV for hydrogen and methane multiplied by heat cost of fuel, OCF.

(4) The fourth step is to choose from the results the best purifier and the best location of the purifier at which the minimum operating cost exists.

(5) The fifth step is to draw the optimum network with the best purifier from the results of the lingo program.

4. Case studies

Two published case studies from the literature and one actual case study were solved to illustrate the ease and applicability of the proposed method.

4.1. Case study 1

Fig. 3 shows a refinery hydrogen network [7,15] where a certain extent of hydrogen integration is included. The existing fresh hydrogen is reported at 277.2 mol/s with 5% [mol%] impurity concentration. The limiting data for this case study are illustrated in Table 1. The purification of this case study is achieved through pressure swing adsorption (PSA) unit with product impurity at 0.1% and hydrogen recovery of 90% or through a membrane unit with product impurity at 2% and a hydrogen recovery of 95% [7,15].

The hydrogen sink flowrate for each hydrotreating process unit is the summation of hydrogen makeup and the recycle of this unit. Also, the hydrogen source flowrate for each hydrotreating process unit is the summation of the recycle and the purge streams of this unit [6]:

1. After applying the first step in the proposed method, it is found that the minimum fresh hydrogen is 268.82 mol/s and the hydrogen discharge is 102.52 mol/s.
2. In case of adding a membrane or a PSA unit to the network and assuming all sources are purified, Table 2 represents the results in the case of adding a membrane unit to the hydrogen network and Table 3 represents the results in the case of adding a PSA unit to the hydrogen network.
3. For each case we calculate the annual operating cost. The cost of production, OCH is taken as \$ 0.075/Nm^3, fuel gas value operating cost is taken as \$ 2.5/MBTU [3,16], and LHV for hydrogen and methane are taken as 229.327 and 760.97 BTU/mol respectively. The operating time is taken as 8000 h/yr. Table 4 represents the operating cost results.
4. From Tables 2 and 3, the optimum design is with the membrane unit when 102.52 mol/s (discharge flowrate) from CNHT at 30% concentration impurity is purified. The minimum fresh hydrogen and fuel discharge are 196.7723 mol/s and 30.4723 mol/s, respectively. The results agree with the previous work [7,15]. As represented in Table 4, source 6 gives the minimum operating cost.
5. Optimum hydrogen network design is shown in Fig. 4.

4.2. Case study 2 (multiple pinch problem)

Table 5 shows the hydrogen sources and sinks data for case study 2 [7] There are six hydrogen sinks and seven hydrogen sources in this network. The purification of this case study is

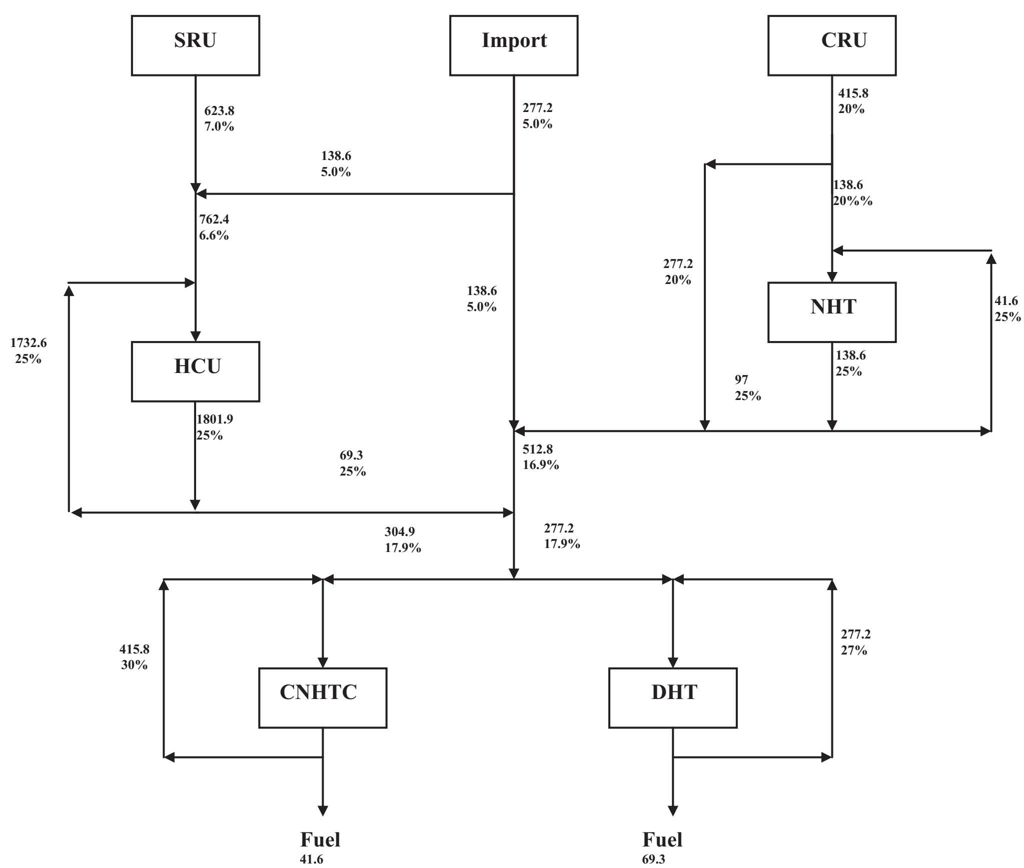

Figure 3 Refinery hydrogen network. The numbers represent the total gas flowrate in (mol/s) and impurity concentration (mol%).

Table 1 Limiting Data for case study 1.

j	Hydrogen sink	Flow rates (mol/s)	Impurity concentration (mol%)
1	HCU	2495.0	19.39
2	NHT	180.2	21.15
3	DHT	554.4	22.43
4	CNHT	720.7	24.86
i	Hydrogen source	Flow rates (mol/s)	Impurity concentration (mol%)
1	SRU	623.8	7.0
2	CRU	415.8	20.0
3	NHT	138.6	25.0
4	HCU	1801.9	25.0
5	DHT	346.5	27.0
6	CNHT	457.4	30.0
	Fresh supply	–	5.0

Table 2 Minimum fresh hydrogen and hydrogen discharge with adding a membrane unit to the network for case study 1.

Cases (with adding a membrane)	Minimum fresh hydrogen (mol/s)	Hydrogen discharge (mol/s)
Source 1 (SRU) is purified	259.6234	93.3234
Source 2 (CRU) is purified	220.7837	54.4837
Source 3 (NHT) is purified	205.8451	39.5450
Source 4 (HCU) is purified	205.8451	39.5450
Source 5 (DHT) is purified	199.8709	33.5709
Source 6 (CNHT) is purified	196.7723	30.4723

Table 3 Minimum fresh hydrogen and hydrogen discharge with adding a PSA unit to the network for case study 1.

Cases (with adding a PSA)	Minimum fresh hydrogen (mol/s)	Hydrogen discharge (mol/s)
Source 1 (SRU) is purified	260.4089	94.1089
Source 2 (CRU) is purified	221.4586	55.1586
Source 3 (NHT) is purified	206.4778	40.1778
Source 4 (HCU) is purified	206.4778	40.1778
Source 5 (DHT) is purified	201.1346	34.8346
Source 6 (CNHT) is purified	199.9884	33.7084

Table 4 Operating cost results for both membrane and PSA units for case study 1.

Cases	Operating cost with adding a membrane unit (*10^6 $/yr)	Operating cost with adding a PSA unit (*10^6 $/yr)
Source 1 (SRU) is purified	9.8694	9.8931
Source 2 (CRU) is purified	8.7046	8.7247
Source 3 (NHT) is purified	8.2566	8.2756
Source 4 (HCU) is purified	8.2566	8.2756
Source 5 (DHT) is purified	8.0733	8.1153
Source 6 (CNHT) is purified	7.9845	8.0765

achieved through a membrane or a PSA unit with properties as described in the previous case study:

1. After applying step 1 of the proposed method, it is found that the minimum fresh hydrogen is 125.2125 mol/s and the hydrogen discharge is 90.9625 mol/s.
2. In this case study, the discharge amount (90.9625 mol/s) is greater than the flowrates of all sources except source 5 (120 mol/s) so we take source 5 to be purified. If all amount of source 5 is taken to be purified as in the previous work [7] and applying the proposed method on the network in the two cases with adding a PSA and with adding membrane unit, Table 6 represents the results.
3. For each case we calculate the annual operating cost. Table 7 represents the operating cost results.
4. As represented in Table 6, it is no profit from the presence of membrane in any case of purification, since the minimum fresh hydrogen and hydrogen discharge of the network with adding a membrane and without adding a membrane are the same. This is identical to the previous work [7]. It is noted also that, the minimum fresh hydrogen and hydrogen discharge are obtained when all of source 5 is purified in a PSA unit.
5. From Table 7, the best purification unit added to the network and giving the minimum operating cost is a PSA unit when all of source 5 is purified.
6. Optimum hydrogen network design is shown in Fig. 5.

4.3. Case study 3: Industrial case study

This case study is representative of a real refinery system. Fig. 6 shows the existing hydrogen network in Midor Refinery Plant at Alexandria-Egypt. There are four consuming units, naphtha hydrotreating, isomerisation, diesel hydrotreating, and hydrocracking unit. The hydrogen is supplied by catalytic reforming unit and hydrogen plant. All the consuming units have recycle compressors except the isomerization unit. Currently 2265.71 kmol/h hydrogen is produced from hydrogen plant. It is noted from Fig. 6 that: the impurity concentration of the recycle stream and the purge stream for naphtha hydrotreating unit (NHT) and hydrocracking unit (HCU) are different, so the recycle and the purge streams flowrates are taken as individual sources. Also, it is noted that the recycle stream for these units is not mixed with the makeup, so the recycle and the makeup streams are taken as individual sinks. Table 8 represents the sink streams and the source streams of the existing network in the refinery.

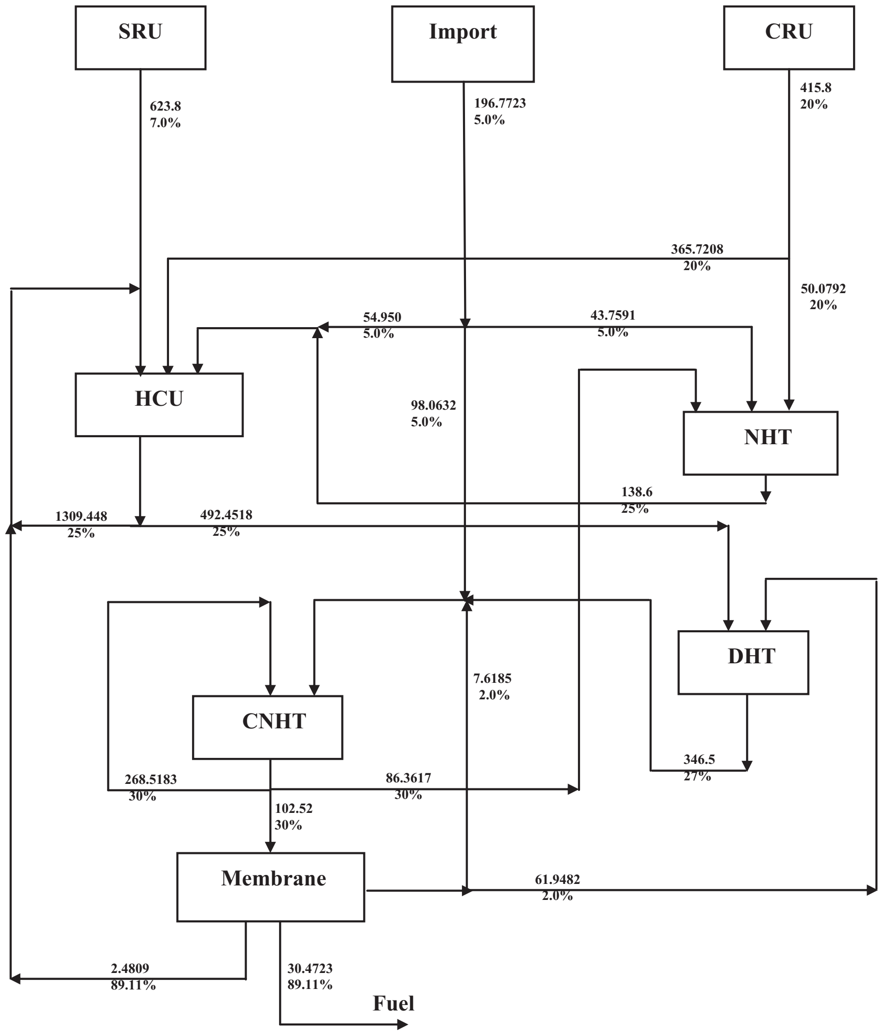

Figure 4 Optimum network design for case study 1. The numbers represent the total gas flowrate in (mol/s) and impurity concentration (mol%).

Table 5 Hydrogen sources and sinks data for case study 2.

	Flow rates (mol/s)	Concentration, Cj (mol%)
Gas sinks SKj		
1	120.0	0.10
2	27.8	1.40
3	60.0	2.50
4	80.0	2.50
5	100.0	3.0
6	150.0	10.0
Gas sources SRi		
1	40.0	1.70
2	80.0	1.70
3	80.0	2.50
4	28.55	4.0
5	80.0	5.0
6	120.0	10.0
7	75.0	15.0
Fresh supply		0.1

There is a PSA unit that is used to purify some amount of hydrogen flow rate from the catalytic reforming unit. The produced purified hydrogen is with 99.9% purity. All purified hydrogen from PSA unit is mixed with the hydrogen produced from hydrogen plant and sent to the hydrocracking unit:

1. After applying step 1 in the proposed method, it is found that: The minimum fresh hydrogen and the discharge are determined to be 2257.964, and 1004.514 kmol/h, respectively.
2. In this case study, all sources flowrates are lower than the discharge flowrate except sources 1 (Outlet 1 NHT), 4 (Outlet DHT), 7 (Outlet 1 HCU), and 9 (CRU). By purifying these sources and using a membrane or a PSA unit, the minimum fresh hydrogen and the discharge for each case are represented in Tables 9 and 10.
3. For each case we calculate the annual operating cost. Table 11 represents the operating cost results.

Table 6 Minimum fresh hydrogen and hydrogen discharge for case study 2.

Cases	Minimum fresh hydrogen (mol/s)	Hydrogen discharge (mol/s)
Network with adding a membrane unit (all source 5 is purified)	125.2125	90.9625
Network with adding a membrane unit (90.96 mol/s from source 5 is purified)	125.2125	90.9625
Network with adding a PSA unit (all source 5 is purified)	54.2308	19.9808
Network with adding a PSA unit (90.96 mol/s from source 5 is purified)	61.2050	26.955

Table 7 Operating cost results for both membrane and PSA units for case study 2.

Cases	Operating cost (*10^6 $/yr)
Network with adding a membrane (all source 5 is purified)	3.748
Network with adding a membrane (90.96 mol/s from source 5 is purified)	3.748
Network with adding a PSA (all source 5, is purified)	1.894
Network with adding a PSA (90.96 mol/s from source 5 is purified)	2.117

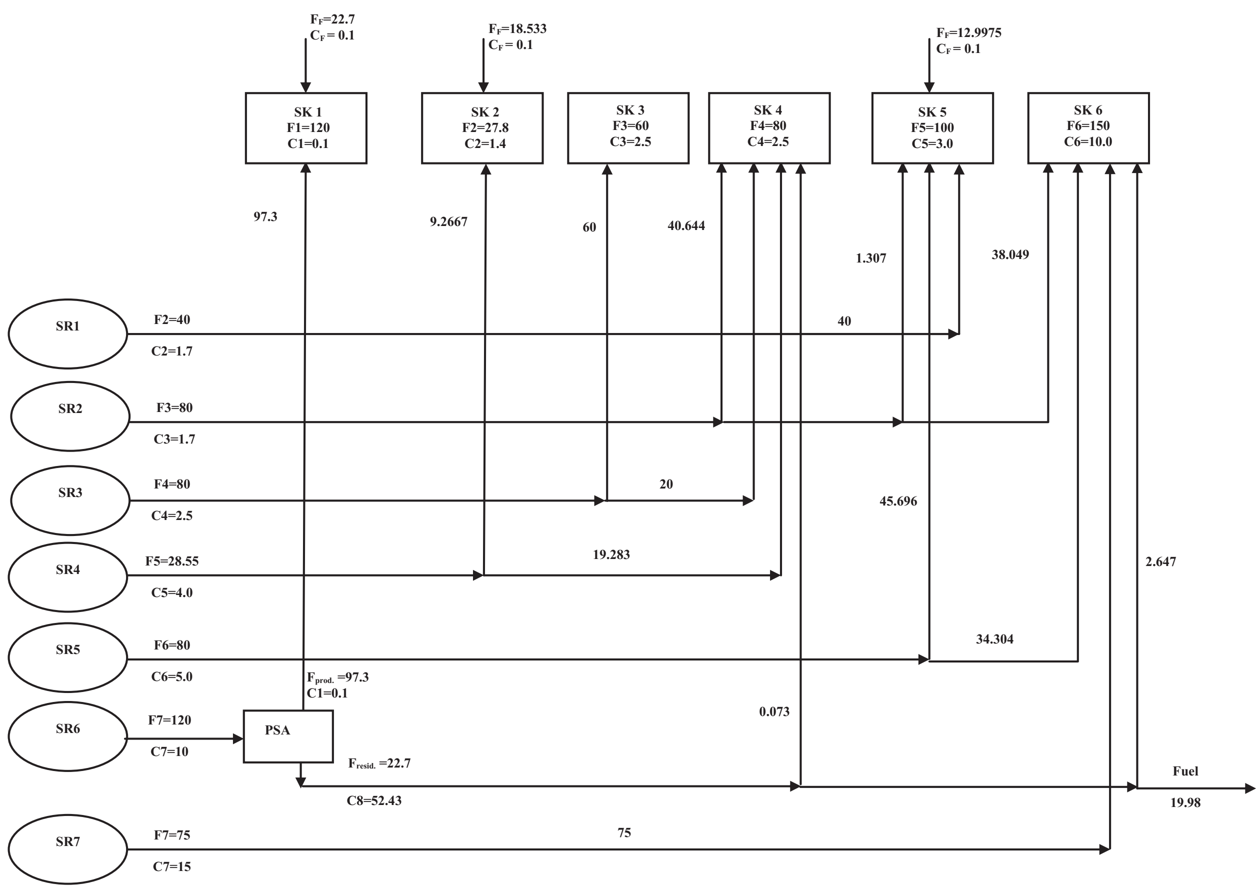

Figure 5 Optimum network design for case study 2. The numbers represent the total gas flowrate in (mol/s) and impurity concentration (mol%).

4. It is noted from Table 9 that the minimum fresh hydrogen and hydrogen discharge are the same when sources 4, 7, and 9 are purified by membrane. In case of using a PSA purifier as represented in Tables 10 and 11, source 4 gives the minimum fresh hydrogen, hydrogen discharge, and the minimum operating cost. The best purification unit added to the network is a PSA unit when an amount equal to the discharge flowrate from source 4 (Outlet DHT) is purified. The fresh hydrogen and discharge flowrate are 1926.361 kmol/h and 672.9111 kmol/h respectively.
5. Optimum hydrogen network design is shown in Fig. 7.

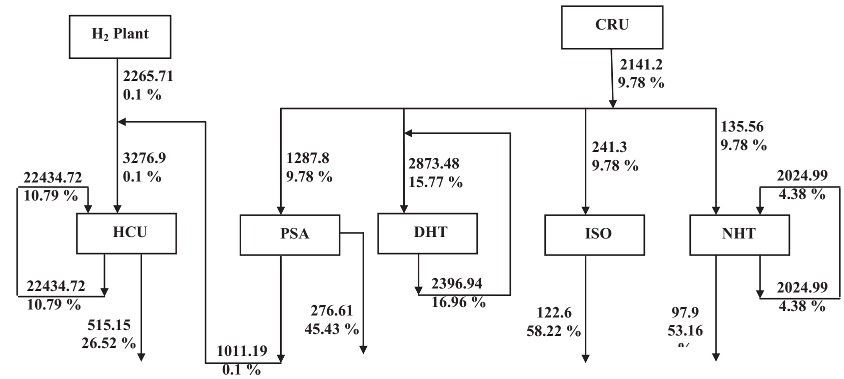

Figure 6 Existing Midor Refinery Plant at Alexandria-Egypt in case study 3 (numbers represent the total gas flowrate in kmol/h and impurity concentration in kmol%).

Table 8 Hydrogen source and sink data for case study 3.

Gas sinks SKj	Hydrogen sink	Flow rates (kmol/h)	Impurity concentration (kmol%)
1	Inlet 1 NHT	2024.99	4.38
2	Inlet 2 NHT	135.56	9.78
3	Inlet ISO	241.3	9.78
4	Inlet DHT	2873.48	15.77
5	Inlet 1 HCU	3276.9	0.1
6	Inlet 2 HCU	22,434.72	10.79
7	Inlet PSA	1287.8	9.78
Gas sources SRi	Hydrogen source	Flow rates (kmol/h)	Impurity concentration (kmol%)
1	Outlet 1 NHT	2024.99	4.38
2	Outlet 2 NHT	97.9	53.16
3	Outlet ISO	122.6	58.22
4	Outlet DHT	2396.94	16.96
5	Outlet 1 PSA	1011.19	0.1
6	Outlet 2 PSA	276.61	45.43
7	Outlet 1 HCU	22,434.72	10.79
8	Outlet 2 HCU	515.15	26.52
9	CRU	2141.2	9.78
	Fresh supply	To be determined	0.01

Table 9 Minimum fresh H_2 and H_2 discharge with adding a membrane unit to the network for case study 3.

Cases (with adding a membrane)	Minimum fresh hydrogen (kmol/h)	Hydrogen discharge (kmol/h)
Source 1 is purified	2235.647	982.197
Source 4 is purified	2163.241	909.791
Source 7 is purified	2163.241	909.791
Source 9 is purified	2163.241	909.791

Table 10 Minimum fresh H_2 and H_2 discharge with adding a PSA unit to the network for case study 3.

Cases (with adding a PSA)	Minimum fresh hydrogen (kmol/h)	Hydrogen discharge (kmol/h)
Source 1 is purified	2234.472	981.022
Source 4 is purified	1926.361	672.911
Source 7 is purified	2049.395	795.945
Source 9 is purified	2078.558	825.108

Table 11 Operating cost results for both membrane and PSA units for case study 3.

Cases	Operating cost with adding a Membrane (*10ˆ6 $/yr)	Operating cost with adding a PSA (*10ˆ6 $/yr)
Source 1 is purified	21.481	21.470
Source 4 is purified	20.839	18.739
Source 7 is purified	20.839	19.831
Source 9 is purified	20.839	21.295

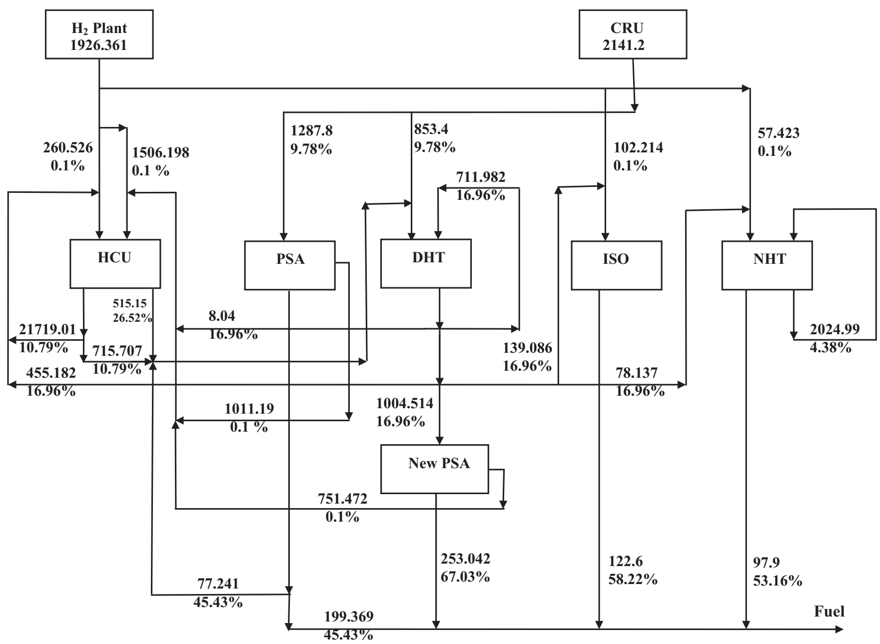

Figure 7 Optimum network design for case study 3. The numbers represent the total gas flowrate in (kmol/h) and impurity concentration (kmol%).

5. Conclusion

A simple proposed optimization method for partitioning regeneration systems is represented. This proposed method gives:

- Accurate identification of the minimum fresh hydrogen and discharge flowrates.
- The appropriate selection of the purifier giving the minimum operating cost.
- Best location of the purifier.

All the solved case studies give the same results as represented in the previous work.

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